

Simple Analytical Expressions of the Steady State Concentration and Flux in Immobilized Glucose Isomerase of Packed - Bed Reactors

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Abstract

The theoretical model of the steady state concentration and flux in immobilized glucose isomerase is discussed. This model is based on diffusion equation containing a non-linear term related to Michaelis-Menten kinetics of the enzymatic reaction. An analytical expression pertaining to the concentration and flux are obtained using the New Homotopy approach for all values of the dimensionless parameters ϕ_p (Thiele modulus) and β (kinetic parameter). Furthermore, in this work the numerical simulation of the problem is also reported using Scilab/Matlab program. The approximate analytical results are compared with the numerical results and found to be in good agreement.

Keywords

Glucose Isomerase; Mathematical Modeling; Reaction Diffusion Equation; Packed Bed; New Homotopy Perturbation Method; Numerical Simulation

Introduction

Pore networks are frequently used in the modeling of transport and reactions in porous materials. This has been reviewed by Sahimi et al.(1990). The equations of diffusion and reaction may be solved by representing the void space with a pore of idealized geometry, usually a cylindrical pore. Marshall and Kooi (1957) discovered glucose isomerase and succeeded in producing it in commercially viable amounts using enzymatic isomerization (EI). Due to this, it was made possible to produce fructose by EI of glucose isomerase [Danno (1970)]. This process was originally carried out in batch reactors with soluble enzymes. It was later extended to one involving immobilized glucose isomerase (IGI). Since with advanced technology large and economical amounts of enzymes can be produced, EI of glucose to fructose using IGI has become one of the most successful enzymatic processes. Melkote and Jensen (1989) used the

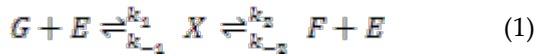
percolation properties of a bethe lattice to model deactivation of glucose isomerase. The effective diffusivity of the pore networks was calculated by Reyes and Jensen (1985). Forment et al (1979, 1982 and 1990) adopted a probabilistic approach to describe the pore network of the catalyst. Mann et al (1986)] simulated fouling within individual catalyst particles using a two dimensional square pore network. Ching and Chu(1988) studied the effect of dispersion and flow on the performance of the reactor. Vasic-Racki et.al (1991) carried out experimental studies and computer modeling of glucose isomerization which was catalyzed by IGI in a packed-bed reactor. Houn et al (1993) carried out experimental studies of the problem using both a differential and a packed-bed reactor and analyzed their own data. They also developed a model for the packed-bed reactor and studied the effect of a variety of factors on the reactors performance. Park et al (1981) and Faqir (1998) investigated the effect of the temperature on the performance of the reactor and developed criteria for optimizing the operating temperature of the reactor.

Recently Dadvar et al (2001) studied deactivation of the micro porous particles as a percolation process by developing a network model. But no general analytical results for the steady state substrate concentration and current for all values of parameters C_{z0} , C_{z1} , ϕ_p and β have been reported. In this paper expressions for the steady state substrate concentration and the current in closed form for small values of parameters is derived using the New Homotopy perturbation approach.

Mathematical Formulations of the Problem and Analysis

In this model we briefly discuss the movement of

glucose enzyme from the intermediate complex form to the fructose and back to the intermediate to the complex glucose enzyme. The reaction can be written as



Here k_1, k_2 are the kinetic constants for the forward direction and k_{-1}, k_{-2} are the kinetic constants for the backward direction. The non-linear reaction diffusion equation for this model can be represented as follows

$$D_p(\lambda) \frac{d^2 G}{dx^2} - \frac{2}{ra} R = 0 \quad (2)$$

where D_p is the pore diffusivity and $\lambda = R_M / r$ (R_M is the molecular radius and r is the pore radius). The non-linear diffusion eqn. (2) is made dimensionless by introducing the following parameters:

$$R = \frac{v_m}{K_m + G} \bar{G}, \quad v_m = \frac{K_{mr} v_{mr} (1 + K^{-1})}{K_{mr} - K_{mf}} \quad (3)$$

$$K_m = \frac{K_{mf} K_{mr}}{K_{mr} - K_{mf}} \left[1 + \left(K_{mf}^{-1} + \frac{K}{K_{mf}} \right) \frac{G_0}{1 + K} \right]$$

The eqn. (2) becomes

$$D_p \frac{d^2 \bar{G}}{dx^2} - \frac{2}{r} \frac{v_m \bar{G}}{K_m + \bar{G}} = 0 \quad (4)$$

Now by introducing the following dimensionless quantities

$$C = \frac{\bar{G}}{(G_0 - G_e)}, \quad z = \frac{x}{l_p}, \quad \beta = \frac{\bar{C}_0}{K_m}, \quad \varphi_p^2 = \frac{2l_p^2 v_m}{r D_p K_m} \quad (5)$$

The eqn.(4) now becomes [17]

$$\frac{d^2 C}{dz^2} - \varphi_p^2 \frac{C}{1 + \beta C} = 0 \quad (6)$$

The boundary conditions can be represented as follows:

$$C = C_{z0} \quad \text{at } z = 0 \quad (7)$$

$$C = C_{z1} \quad \text{at } z = 1 \quad (8)$$

The dimensionless current is given by

$$J_{ij} = \left[\frac{\partial C}{\partial z} \right]_{z=1} \quad (9)$$

Analytical Expressions of the Concentration and Current using the New Homotopy Perturbation Method

Recently, non-linear differential equations are solved by various analytical methods. Lesnic (2007) used the Adomian decomposition method. He (2006), Ozis et.al (2007) solved by Homotopy perturbation method and Zhu (2007) applied exp-function method. The Homotopy algorithm might be improved, if Rentoul and Ariel algorithm is adopted (2011). The HPM is unique in its applicability, accuracy and efficiency. The HPM uses the imbedding parameter p as a small parameter, and only a few iterations are needed to search for an asymptotic solution. This method is the most effective and convenient ones for both linear and non-linear equations. Perturbation method is based on assuming a small parameter. The majority of non-linear problems, especially those having strong non-linearity, have no small parameters at all and the approximate solutions obtained by the perturbation methods, in most cases, are valid only for small values of the small parameter. The New Homotopy perturbation method [Rajendran and Ananthaswamy et. al. (2012, 2013 and 2014)] is very simple and effective. Using this method, we obtain the analytical expression of substrate concentration (see Appendix B) as follows:

$$C(z) = \frac{C_{z0} \sinh\left(\frac{\varphi_p (1-z)}{\sqrt{1+\beta C_{z0}}}\right) + C_{z1} \sinh\left(\frac{\varphi_p z}{\sqrt{1+\beta C_{z0}}}\right)}{\sinh\left(\frac{\varphi_p}{\sqrt{1+\beta C_{z0}}}\right)} \quad (10)$$

The eqn.(10) is the new analytical expression for the dimensionless substrate concentration in terms of the dimensionless kinetic parameter β and the Thiele modulus φ_p . The current density by using the eqn.(9) is,

$$J_{ij} = \frac{C_{z1} \cosh\left(\frac{\varphi_p z}{\sqrt{1+\beta C_{z0}}}\right) \left(\frac{\varphi_p}{\sqrt{1+\beta C_{z0}}}\right) - C_{z0} \cosh\left(\frac{\varphi_p (1-z)}{\sqrt{1+\beta C_{z0}}}\right) \left(\frac{\varphi_p}{\sqrt{1+\beta C_{z0}}}\right)}{\sinh\left(\frac{\varphi_p}{\sqrt{1+\beta C_{z0}}}\right)} \quad (11)$$

Numerical Simulation

The non-linear differential equations (6)-(8) are also solved numerically. We have used the function main in Matlab/Scilab software to solve the initial-boundary value problems for the non-linear differential equations numerically. This numerical solution is compared with our analytical results in Figures 1 -2. Upon comparison, it gives a satisfactory agreement for all values of the dimensionless parameters C_{z0} , C_{z1} , ϕ_p and β .

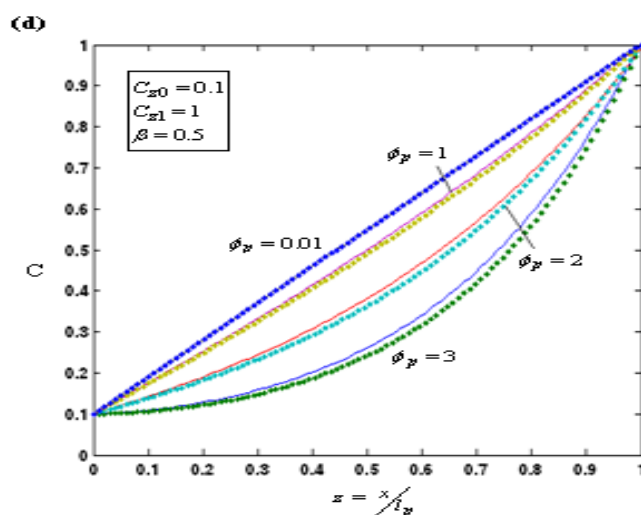
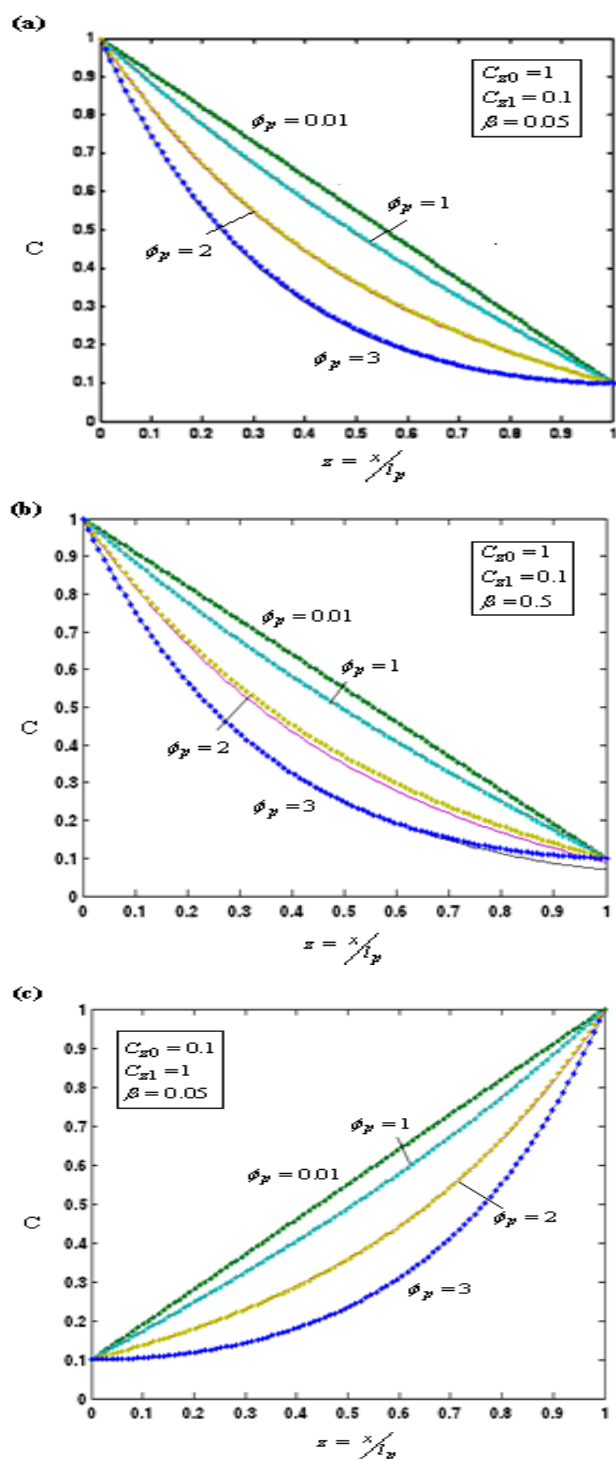
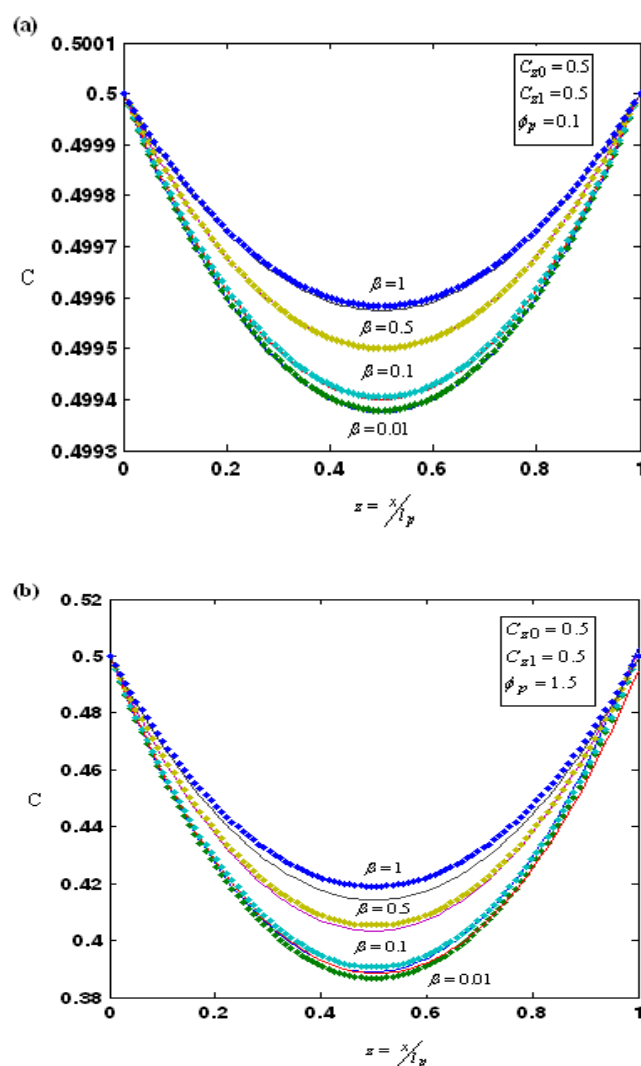


FIGURE 1 (A-D): PLOT OF THE CONCENTRATION C VERSUS THE NORMALIZED DISTANCE z FOR DIFFERENT VALUES OF ϕ_p AND SOME FIXED VALUES OF C_{z0} , C_{z1} AND β . THE CONCENTRATIONS WERE COMPUTED USING THE EQN.(10). THE KEY OF THE GRAPH: (-) REPRESENTS THE EQN.(10) AND (*) REPRESENTS THE NUMERICAL SIMULATION.



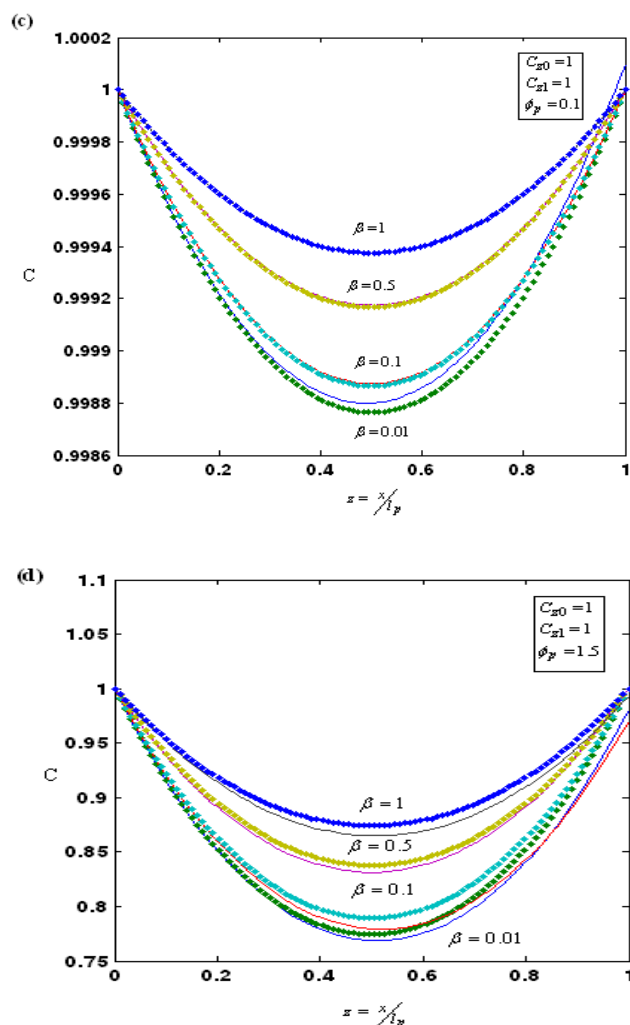
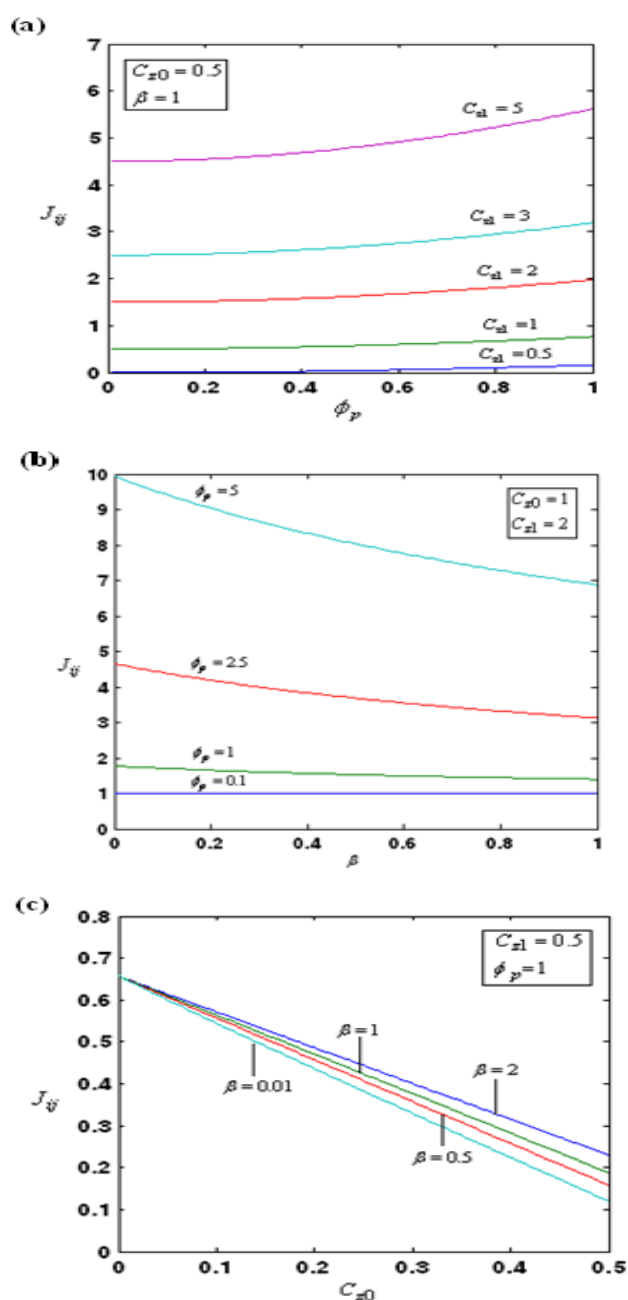


FIGURE 2(A-D): PLOT OF THE CONCENTRATION C VERSUS THE NORMALIZED DISTANCE z FOR DIFFERENT VALUES OF β AND SOME FIXED VALUES OF C_{z0} , C_{z1} AND ϕ_p . THE CONCENTRATIONS WERE COMPUTED USING THE EQN.(10). THE KEY OF THE GRAPH: (-) REPRESENTS THE EQN.(10) AND (*) REPRESENTS THE NUMERICAL SIMULATION.

Results and Discussions

The eqn.(10) represent the closed and simple approximate analytical expression of the concentration of substrate for small values of parameters C_{z0} , C_{z1} , ϕ_p and β . The eqn.(11) represent new simple analytical expression of flux. The concentration of substrate depends upon the values of the parameters C_{z0} , C_{z1} , ϕ_p and β . The parameter Thiele modulus ϕ_p can be varied by changing either the thickness of the enzyme layer or the amount of enzyme immobilized in the matrix. This parameter describes the relative importance of diffusion and reaction in the enzyme layer. Fig. 1 and 2 represents the dimensionless steady state concentration C for different values of the thiele modulus ϕ_p and

dimensionless kinetic parameter β . From Fig.(2), it is obvious that when the Thiele modulus ϕ_p increases, the corresponding substrate concentration C decreases in some fixed values of the other dimensionless parameters. From Fig.(2), it is observed that, when dimensionless kinetic parameter β increases, the corresponding substrate concentration C also increases in some fixed values of the other dimensionless parameters. The Fig. 3(a-d) represents the dimensionless flux J_{ij} versus dimensionless kinetic parameter β for various values of Thiele modulus ϕ_p . From this Fig. it is evident that the value of the flux increases as the dimensionless kinetic parameter β and the Thiele modulus ϕ_p increases.



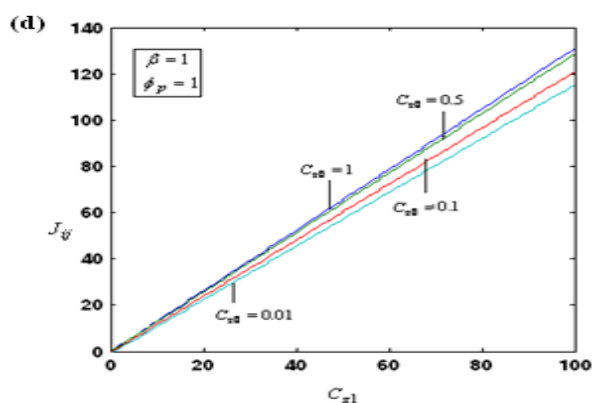


FIGURE:3 VARIATION OF FLUX J_{ij} VERSUS DIMENSIONLESS PARAMETER (A) ϕ_p (B) β (C) C_{z0} AND (D) C_{z1} . CURRENT IS COMPUTED USING THE EQN.(11).

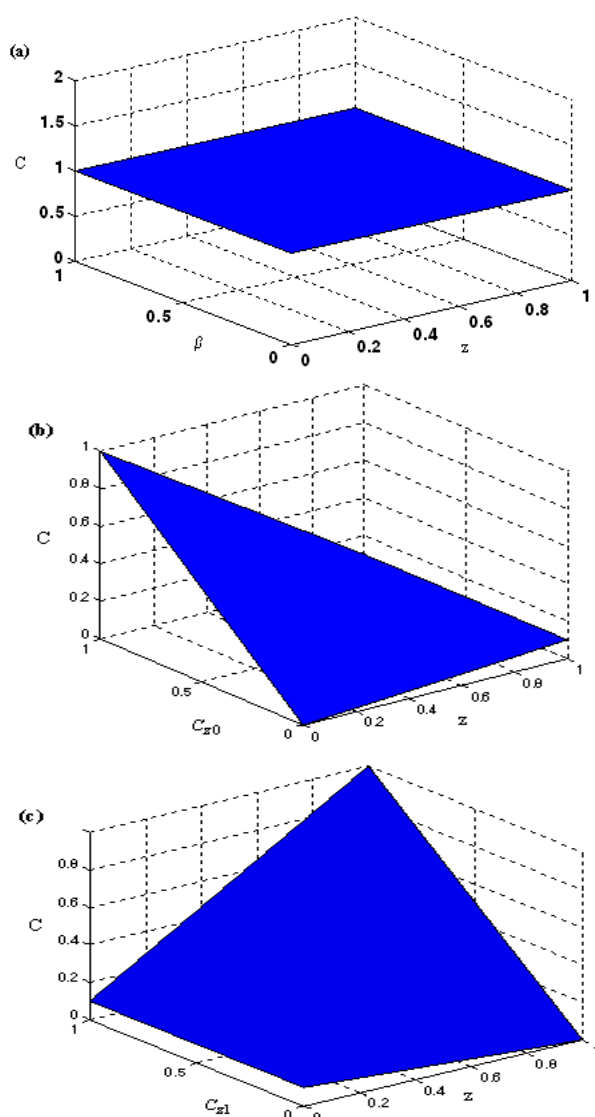


FIGURE: 4 THE NORMALIZED THREE-DIMENSIONAL NORMALIZED CONCENTRATION PROFILE THE EQN.(10) (A) WHEN $0 \leq \beta \leq 1$, $\phi_p = 0.01$, $C_{z0} = 1$ AND $C_{z1} = 1$ (B) WHEN $0 \leq C_{z0} \leq 1$, $\beta = 0.01$, $\phi_p = 1$ AND $C_{z1} = 0.1$ (C) WHEN $0 \leq C_{z1} \leq 1$, $\beta = 0.01$, $\phi_p = 1$ AND $C_{z0} = 0.1$.

Conclusions

The steady state concentration and flux in immobilized glucose isomerase of packed bed reactors which exhibits Michaelis Menten constant has been discussed. The approximate analytical expressions of the non linear reaction diffusion equation have been derived. Analytical expressions for the concentration and flux in packed bed reactor for kinetic reactions with diffusion coefficients at a planar microelectrode under steady state conditions are obtained using the New Homotopy approach. The primary result of this work is simple approximate calculation of concentration profiles and flux for small values of the fundamental parameters. The small variation caused a significant change in both the magnitude of the current and the general behavior of the system. The New Homotopy approach is very simple to solve the non-linear equations. This method can be easily extended to find the solution of all other non linear reaction diffusion equations in kinetic reactions for various complex boundary conditions.

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Author Introduction



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Appendix A: Basic Concept of the Homotopy Perturbation Method

To explain this method, let us consider the following function:

$$D_o(u) - f(r) = 0, \quad r \in \Omega \quad (\text{A.1})$$

with the boundary conditions of

$$B_o(u, \frac{\partial u}{\partial n}) = 0, \quad r \in \Gamma \quad (\text{A.2})$$

where D_o is a general differential operator, B_o is a boundary operator, $f(r)$ is a known analytical function and Γ is the boundary of the domain Ω . In general, the operator D_o can be divided into a linear part L and a non-linear part N . The eqn.(A.1) can therefore be written as

$$L(u) + N(u) - f(r) = 0 \quad (\text{A.3})$$

By the Homotopy technique, we construct a Homotopy $v(r, p) : \Omega \times [0, 1] \rightarrow \Re$ that satisfies

$$H(v, p) = (1 - p)[L(v) - L(u_0)] + p[D_o(v) - f(r)] = 0 \quad (\text{A.4})$$

$$H(v, p) = L(v) - L(u_0) + p[L(u_0) + p[N(v) - f(r)]] = 0 \quad (\text{A.5})$$

where $p \in [0, 1]$ is an embedding parameter, and u_0 is an initial approximation of the eqn.(A.1) that satisfies the boundary conditions. From the eqns. (A.4) and (A.5), we have

$$H(v, 0) = L(v) - L(u_0) = 0 \quad (\text{A.6})$$

$$H(v, 1) = D_o(v) - f(r) = 0 \quad (\text{A.7})$$

When $p=0$, the eqns.(A.4) and (A.5) become linear equations. When $p=1$, they become non-linear equations. The process of changing p from zero to unity is that of $L(v) - L(u_0) = 0$ to $D_o(v) - f(r) = 0$. We first use the embedding parameter p as a small parameter and assume that the solutions of the eqns. (A.4) and (A.5) can be written as a power series in p :

$$v = v_0 + pv_1 + p^2v_2 + \dots \quad (\text{A.8})$$

TABLE: 1 COMPARISON OF THE ANALYTICAL RESULT WITH MARGERET ET AL WITH THE NUMERICAL RESULT OF THE CONCENTRATION C
WHEN $\varphi_p = 2$, $C_{z_0} = 0.5$, $C_{z_1} = 1$ AND FOR VARIOUS VALUES OF β

z	$\beta = 0.01$			$\beta = 1$			$\beta = 2.5$			$\beta = 5$		
	Numeric al	This work	Margeret et al	Numeric al	This work	Margeret et al	Numerica l	This work	Margeret et al	Numerical	This work	Margeret et al
0	0.5000	0.5000(0.00)	(0.00)	0.5000	0.5000(0.00)	(0.02)	0.5000	0.5000(0.00)	(0.00)	0.5000	0.5000(0.00)	(0.00)
0.2	0.4411	0.4413(0.05)	(0.63)	0.4862	0.4825(0.77)	(1.08)	0.5238	0.5157(1.57)	(2.98)	0.5521	0.5427(1.73)	(13.4)
0.4	0.4548	0.4538(0.22)	(0.93)	0.5262	0.5170(1.78)	(4.26)	0.5847	0.5682(2.90)	(1.22)	0.6281	0.6102(2.93)	(10.3)
0.6	0.5371	0.5395(0.44)	(0.80)	0.6165	0.6071(1.55)	(6.83)	0.6785	0.6615(2.57)	(6.39)	0.7233	0.7058(2.48)	(1.36)
0.8	0.7102	0.7123(0.29)	(0.44)	0.7726	0.7625(1.32)	(5.99)	0.8173	0.8021(1.90)	(7.50)	0.8484	0.8338(1.75)	(5.84)
1	1.0000	1.0000(0.00)	(0.00)	1.0000	1.0000(0.00)	(0.00)	1.0000	1.0000(0.00)	(0.00)	1.0000	1.0000(0.00)	(0.00)
Average deviation		0.17	0.47		0.90	3.03		1.49	3.02		1.48	5.15

TABLE: 2 COMPARISON OF THE ANALYTICAL RESULT WITH MARGERET ET AL WITH THE NUMERICAL RESULT OF THE CONCENTRATION C
WHEN $\beta = 0.01$, $C_{z_0} = 0.1$, $C_{z_1} = 1$ AND FOR VARIOUS VALUES OF φ_p

z	$\varphi_p = 0.01$			$\varphi_p = 0.1$			$\varphi_p = 1$			$\varphi_p = 3$		
	Numeric al	This work	Margeret et al	Numerica l	This work	Margeret et al	Numerica l	This work	Margeret et al	Numerical	This work	Margeret et al
0	0.1000	0.1000(0.00)	(0.00)	0.1000	0.1000(0.00)	(0.00)	0.1000	0.1000(0.00)	(0.00)	0.1000	0.1000(0.00)	(0.00)
0.2	0.2818	0.2800(0.64)	(0.04)	0.2814	0.2796(0.64)	(0.25)	0.2486	0.2469(0.69)	(3.26)	0.1187	0.1182(0.42)	(3.81)
0.4	0.4436	0.4600(0.78)	(0.24)	0.4630	0.4594(0.78)	(0.22)	0.4073	0.4037(0.89)	(0.04)	0.1823	0.1802(1.17)	(4.39)
0.6	0.6364	0.6400(0.56)	(0.03)	0.6357	0.6393(0.56)	(0.16)	0.5733	0.5765(0.59)	(2.25)	0.3059	0.3089(0.97)	(3.27)
0.8	0.8182	0.8200(0.22)	(0.01)	0.8177	0.8195(0.22)	(0.09)	0.7710	0.7729(0.25)	(1.22)	0.5595	0.5522(0.49)	(1.67)
1	1.0000	1.0000(0.00)	(0.00)	1.0000	1.0000(0.00)	(0.00)	1.0000	1.0000(0.00)	(0.00)	1.0000	1.0000(0.00)	(0.00)
Average deviation		0.37	0.05		0.37	0.12		0.40	1.13		0.51	2.19

Setting $p = 1$ results in the approximate solution of the eqn. (A.1):

$$u = \lim_{p \rightarrow 1} v = v_0 + v_1 + v_2 + \dots \quad (\text{A.9})$$

This is the basic idea of the HPM.

Appendix B: Solution of the Non-linear Differential Eqns.(6)-(8) Using the New Homotopy Perturbation Method

In this Appendix, we indicate how the eqn.(10) in this paper is derived. To find the solution of the eqn.(10), we first construct a Homotopy as follows:

$$(1-p) \left[\frac{d^2 C}{dz^2} - \varphi_p^2 C \right] + p \left[(1+\beta C) \frac{d^2 C}{dz^2} - \varphi_p^2 C \right] = 0 \quad (\text{B.1})$$

The approximate analytical solution of the eqn.(B.1) is

$$C = C_0 + pC_1 + p^2C_2 + p^3C_3 + \dots \quad (\text{B.2})$$

Substituting the eqn. (B.2) into an eqn.(B.1) we get

$$(1-p) \left[\frac{d^2 (C_0 + pC_1 + p^2C_2 + p^3C_3 + \dots)}{dz^2} - \varphi_p^2 (C_0 + pC_1 + p^2C_2 + p^3C_3 + \dots) \right] + p \left[\frac{d^2 (C_0 + pC_1 + p^2C_2 + p^3C_3 + \dots)}{dz^2} - \varphi_p^2 (C_0 + pC_1 + p^2C_2 + p^3C_3 + \dots) \right] = 0 \quad (\text{B.3})$$

The initial approximations are as follows:

$$C_0(0) = C_{z_0}; C_0(1) = C_{z_1} \quad (\text{B.4})$$

$$C_i(0) = 0; C_i(1) = 0, i = 1, 2, 3, \dots \quad (\text{B.5})$$

Comparing the coefficients of like powers of p in eqn. (B.3) we get

$$p^0: \frac{d^2 C_0}{dz^2} - \frac{\varphi_p^2 C_0}{(1+\beta C_{z_0})} = 0 \quad (\text{B.6})$$

Solving the eqn. (B.6) and using the initial

approximation eqn. (B.4) we can obtain the following result:

$$C_0 = \frac{C_{z0} \sinh\left(\frac{\phi_p(1-z)}{\sqrt{1+\beta C_{z0}}}\right) + C_{z1} \sinh\left(\frac{\phi_p z}{\sqrt{1+\beta C_{z0}}}\right)}{\sinh\left(\frac{\phi_p}{\sqrt{1+\beta C_{z0}}}\right)} \quad (\text{B.7})$$

According to the Homotopy perturbation method, we can conclude that

$$C = \lim_{p \rightarrow 1} C(z) = C_0 \quad (\text{B.8})$$

Substituting the eqn.(B.7) into an eqn. (B.8), we can obtain the solution in the text.

Appendix C: Nomenclature

Symbol	Meaning
G	Reaction of glucose
E	Reaction of enzyme
F	Reaction of fructose
G_0	Initial concentration
D_p	Pore Diffusivity
R	Reaction rate
X	Complex intermediate
K_{mf}	Michaelis-Menten constant
v_{mf}	Maximum velocity of the forward reaction
K_{mr} and v_{mr}	Maximum velocity of the backward reaction
C	Dimensionless concentration
l_p	Pore length
ϕ_p	Pore level Thiele-modulus
C_{z0}, C_{z1}	Constants
J_{ij}	Flux
k_1, k_2, k_{-1}, k_{-2}	Kinetic constants